

GASTROINTESTINAL ABSORPTION OF PLUTONIUM BY THE MARSHALL ISLANDERS

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Abstract—The gastrointestinal absorption constant (f_1) is a critical parameter in assessing systemic uptake following the ingestion of a radioactive material and in monitoring such intakes. This study addresses the latter, particularly for plutonium, and from environmental measurements derives an f_1 value of 4×10^{-4} for the Marshallese population. The uncertainty associated with the methodology and measurements used in this f_1 value assessment is evaluated. This evaluation takes into account the results from 24-h urine samples and the particular lifestyle of the Marshallese. Plutonium intake resulting from soil consumption is a primary parameter in this evaluation; for this study, it was assumed to be 500 mg d^{-1} . The f_1 value determined here is consistent with the values in ICRP Publication 67 of 5×10^{-4} for ages 1 to adult, and is the same as that suggested by the NRPB. *Health Phys.* 73(1):167–175; 1997

Key words: gastrointestinal tract; Marshall Islands; plutonium; soil

INTRODUCTION

THE INTAKE of radioactive material can occur principally through three major pathways: inhalation, ingestion, and absorption through open wounds. Uptake is the fraction of the intake that reaches the systemic system of the body through any of these pathways. In this study, the quantity of plutonium entering the gastrointestinal (GI) tract is limited to ingestion, and since the uptake is considered to be proportional to the fraction of the ingested plutonium that is absorbed through the gut wall into the blood stream, it can be expressed as a constant: the gastrointestinal absorption coefficient (f_1). Therefore, for ingestion, the systemic uptake via the GI tract is the product of the intake and the f_1 value.

The International Commission on Radiological Protection (ICRP) in Publication 30 (1978) introduced the annual limit on intake (ALI) and provided dose per unit intake coefficients for controlling occupational exposure to internally deposited radionuclides. In that publication, the intake of one ALI, either from inhalation or ingestion, was taken to result in a committed effective dose of 50

mSv in the 50-y interval following an intake. Inherent in calculating ALIs by ingestion is the f_1 value. In ICRP Publication 48 (1986) f_1 value of 10^{-3} was recommended for adults for unknown or mixed compounds with the intention of providing “an adequate margin of safety for radiological protection purposes” (ICRP 1987). However, they suggested a value of 10^{-2} for infants for the first year of life. Similar values of 10^{-2} for infants and 10^{-3} for adults were also used in Publication 56 for plutonium in the diet (ICRP 1989). Both ICRP Publication 67 (1993) and the U.K. National Radiological Protection Board Gut Transfer Report (NRPB 1990) gave values of 5×10^{-3} for infants and 5×10^{-4} for adults.

As Kocher and Ryan (1983) indicated, these values are intended to be used for dose limitation. Durbin (1975) indicated the plutonium absorption from the GI tract, wound sites, or the lung decreases in the following order: soluble complexes > hydrolyzable salts > insoluble compounds of plutonium. It is important to remember, however, as stated in ICRP Publication 48 (ICRP 1986) “the use of the cautious value of 10^{-3} may not be considered appropriate in all situations where a best estimate of absorption is required, either for a critical group or in estimating a population dose.” Underestimating the f_1 value would increase the estimate of the intake as interpreted from urinalysis. It is for this reason that we undertook an examination of the most appropriate f_1 value for the Marshallese people.

Other studies

There have been numerous studies on the behavior of plutonium in human and animal biological systems. In a study in which plutonium was fed chronically to rats, Weeks et al. (1956) determined an f_1 value of about 3×10^{-3} . On the other hand, an f_1 value of $\sim 10^{-5}$ has been cited by Priest and Tasker (1990). Pinder et al. (1990) suggested f_1 values of 10^{-3} and 10^{-5} for the ingestion of plutonium from plants which incorporated plutonium via roots from contaminated soil and from plants with surface contamination, respectively. Bhattacharyya et al. (1992) used Pu(+4) and found that GI absorption values were similar in mice, baboons, and humans and they suggested an f_1 value of 1×10^{-4} . From their investigation of plutonium levels in urine from people whose diet included shellfish, Hunt et al. (1990)

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proposed an f_1 value of 2×10^{-4} from Cumbrian winkles. Another similar study based on the ingestion of plutonium incorporated in reindeer meat concluded that the f_1 value is about 8×10^{-4} (Mussalo-Rauhamaa et al. 1984). Between April 1945 and July 1947, Pu(+4) and Pu(+6) citrate solutions were selected and injected in ill men to study plutonium toxicity in humans (Russell 1946; Russell and Nickson 1946; Langham et al. 1980). Also, Popplewell et al. (1994) carried out experiments with $^{244}\text{Pu}(+4)$ in volunteers and suggested an f_1 value between 2×10^{-4} and 9×10^{-4} for their human biokinetics modeling of plutonium. Later, Langham's results (Langham et al. 1980) were thoroughly reviewed and the data extrapolated in the development of biokinetic models for dose calculations and for interpretation of bioassay data (Beach and Dolphin 1964; Durbin 1975; Rundo et al. 1976; Parkinson and Henley 1981; Leggett 1984; Jones 1985; Kathren and McInroy 1991; McInroy et al. 1991; Voelz and Lawrence 1991; Moss and Eckhardt 1995).

Taylor (1989) suggested that the f_1 value for plutonium and other actinides and transuranium elements could vary by three orders of magnitude depending upon the mass ingested, the specific compounds ingested, and the individual's dietary habits and physiology. Smith et al. (1975) indicated that smaller particle sizes give lower f_1 values due to the greater retention time in the GI tract. On the other hand, Sullivan (1980a, b) indicated that smaller particle sizes give larger f_1 values due to easier digestion in the GI tract. Larsen et al. (1977, 1983) suggested a f_1 value for plutonium between 10^{-3} and 2×10^{-1} based on the chemistry of plutonium in chlorinated drinking water. However, animal studies have not produced convincing evidence that either the oxidation state of the ingested compound or individual biology influence the f_1 value (NEA 1988). Uptake is proportional to plutonium intake, and each individual's intake differs, as do living habits. Therefore, a proper and correct f_1 value must take into account an individual's living habits, such as foods consumed (and their preparation) (McKay and Fox 1991; ICRP 1989).

In a study on plutonium retention in the intestinal wall of rats, the f_1 value for older animals was found to be about 10% of that for younger animals (NRPB 1990; Harrison and Fritsch 1992). This finding is consistent with the ICRP Publication 67 (ICRP 1993) recommended f_1 values of 5×10^{-3} for 3-mo-old infants and 5×10^{-4} for those aged 1 y and older. Therefore, it is more likely that the f_1 value remains nearly constant for variables except age, and perhaps gender, and it is variable intake which leads to differences in excretion. Moreover, the current plutonium retention-excretion models and biokinetics parameter values (ICRP 1989, 1993) allow for age-dependent changes in bone physiology, which are reflected in changes in urine excretion as a function of age. Therefore, the uncertainty of an assessed f_1 value for Marshallese populations using urine data may be overlooked.

MATERIALS AND METHODS

Data obtained through urine measurements

On the morning of 1 March 1954, a device code named Bravo was detonated at Namu Island, Bikini Atoll, the Republic of Marshall Islands (RMI). An unexpectedly large yield resulted in radioactive fallout inadvertently contaminating two inhabited atolls, Rongelap and Utirik (U.S. Committee on Interior and Insular Affairs 1989; U.S. Committee on Energy and Natural Resources 1991; National Research Council 1994; U.S. Committee on Natural Resources 1994). The current work is limited primarily to the Rongelap population.

The Bravo test released plutonium isotopes that were transported to Rongelap and Utirik in fallout. All 64 Rongelap residents were evacuated 2 d after the Bravo detonation. These residents, therefore, were exposed to acute radiation from early fallout both before and during their evacuation. In June 1957, the whole population was allowed to return to Rongelap Atoll. However, in May 1985, the entire community was evacuated again due to fear of exposure to fallout radiation. At present, no one lives in the Rongelap Atoll. Thus, from June 1957 to May 1985, the original 64 inhabitants and their descendants were chronically exposed to low-level plutonium contamination while living on the Atoll. In the summer of 1989, a BNL mission conducted whole-body counting and collected urine samples from 34 Rongelapese living at Mejjatto Island (Sun et al. 1992, 1995). The estimated number of Rongelap people living at Mejjatto then was 200–250, with half being children under the age of 10 y. After May 1985, while living on Mejjatto, the Rongelap people were unlikely to ingest any plutonium as evidenced by the low levels of radiocesium activity detected there. Our interpretation of the urine data is based on these chronological events, and, therefore, assumes that no intake of plutonium occurred before March 1954 nor after May 1985.

In 1989, the fission track analysis (FTA) method was fully implemented (Moorthy et al. 1988) and a protocol established for a 24-h shipboard urine collection that led to a set of reliable data on ^{239}Pu excretion in urine (Sun et al. 1993a). For example, the average plutonium body content obtained from 24-h urine samples is in good agreement with that from analyses of bone samples of deceased Rongelap residents (Franke et al. 1995).

Calculation method and measurements

Table 1 shows the ages, sex, and the time of residence on the Rongelap atoll of the 34 people, together with their individual FTA results. The results are indicated as gross and net plutonium contents (μBq) in 24-h urine samples. The average total background from reagent urine blanks and systematic background is about $1.6 \mu\text{Bq}$ [36 fission tracks with each fission track equivalent to $0.044 \mu\text{Bq}$ (Sun et al. 1995)]. In order to obtain a positive result of plutonium in the body, the gross plutonium content in the sample must be no less

Table 1. Estimates of ^{239}Pu activity in 24-h urine samples and total days lived on Rongelap Island from 34 Rongelap people.

ID	Age ^a	Sex	^{239}Pu gross activity in 24-h urine (μBq)	^{239}Pu net activity in 24-h urine (μBq)	Exposure days ^b
1	19	M	4.5	2.9	5,475
2	14	M	3.4	1.8	3,650
3	12	M	2.9	1.3	2,920
4	14	M	3.2	1.6	3,650
5	16	M	3.4	1.8	4,380
6	16	M	3.4	1.8	4,380
7	15	M	3.0	1.4	4,015
8	12	M	2.3	0.7	2,920
9	14	M	2.2	0.6	3,650
10	16	M	2.2	0.6	4,380
11	14	M	1.9	0.3	3,650
12	10	M	1.7	0.1	2,190
13	19	M	1.7	0.1	5,475
14	10	F	3.0	1.4	2,190
15	15	F	3.2	1.6	4,015
16	11	F	2.4	0.8	2,555
17	10	F	2.3	0.7	2,190
18	11	F	2.3	0.7	2,555
19	15	F	2.7	1.1	4,015
20	85	F	4.5	2.9	10,220
21	16	F	2.3	0.7	4,380
22	13	F	1.9	0.3	3,285
23	15	F	2.0	0.4	4,015
24	10	F	1.7	0.1	2,190
25	12	F	1.6	0	2,920
26	17	F	1.6	0	4,745
27	33	F	1.1	-0.5	10,220
28	14	F	0.8	-0.8	3,650
29	14	F	1.4	-0.2	3,650
30	14	F	1.5	-0.1	3,650
31	16	F	1.2	-0.4	4,380
32	12	F	1.3	-0.3	2,920
33	13	F	1.2	-0.4	3,285
34	66	F	1.1	-0.5	10,220
Total			76.4	22.5	141,985

^a The age during the year of sample collection.^b Days calculated between 16 June 1957 and 15 May 1985.

than $1.6 \mu\text{Bq}$. However, 8 of the 34 individuals' sample results were shown below $1.6 \mu\text{Bq}$ and resulted net plutonium contents in a negative value. Clearly, a negative plutonium content in the urine is not possible, nor is a negative f_1 value, as might be suggested from these 8 individual negative net plutonium results. To avoid nonsensical statistical biases, $22.5 \mu\text{Bq}$ plutonium was obtained from the sum of the 34 FTA gross results ($76.4 \mu\text{Bq}$) then subtracted by total background of the 34 FTA analyses ($1.6 \mu\text{Bq} \times 34 = 53.9 \mu\text{Bq}$). Similarly, 141,985 days were simply summed from the same 34 people who lived in the Rongelap Islands between June 1957 and May 1985. These two totals are shown on the bottom line of Table 1.

The population examined in Table 1 has an age range between 10 and 85 y. Since the plutonium elimination rate is faster in older individuals, the fractional daily excretion might be expected to differ between individuals in this population. Priest and Birchall (1989) reported that the ICRP age-specific model for bone-surface seeking radionuclides in humans is relatively insensitive to the age of the individuals, and the pluto-

nium urine excretion model applied to the adult age group still provides sufficient accuracy for estimation of intake for all ages except infants. Therefore, the variation of plutonium excretion rates between age 10 to 85 y is small as suggested by the ICRP (1989, 1993) human bone model, and translocation parameter values among bone, blood, urinary tract tissue, and urine compartments are small as well. Hence, based on Jones's plutonium excretion model (1985), a predicted fractional elimination of 1.4×10^{-5} of an original single acute uptake can be applied for the following 4 y. Therefore, a total plutonium uptake of 1.61 Bq (i.e., $22.5 \mu\text{Bq} \div 1.4 \times 10^{-5}$) would be interpreted as the average plutonium body content of the 34 people in May 1985 when they left Rongelap Island. Further, based on the suggestion in ICRP Publication 67 (1993) that about 80% of the plutonium uptake absorbed in the systemic whole-body is available for a rapid elimination, the total uptake (systemic burden) of ^{239}Pu is about 2.01 Bq (i.e., $1.61 \text{ Bq} \div 0.8$). Therefore, the average rate of plutonium uptake for these 34 individuals from both the inhalation

and ingestion pathways was about $14.2 \mu\text{Bq d}^{-1}$ (i.e., $2.01 \text{ Bq} \div 141,985 \text{ d}$).

An alternative calculation can be made using non-parametric statistics. For FTA data generated during 1989, the minimum detection level (MDL) was $2 \mu\text{Bq}$ (99% confidence level). It is seen in Table 1 that a large number of results were below this value. As reported by Helsel (1990), there are several statistical procedures for handling values reported as less-than MDLs, such as substituting zero for such values, or one-half the MDL, or even the MDL itself. However, each method introduces bias when estimates of means and variances for the population are made. An alternative is to use percentiles (e.g., medians, interquartile range), which are robust parameter estimators for entire population data sets. Using all 34 net plutonium results in column 5, Table 1, the median value is $0.6 \mu\text{Bq}$ (e.g., compared to the mean value, $0.66 \mu\text{Bq}$). This translates into an intake of $12.8 \mu\text{Bq d}^{-1}$, which is only 10% below the previous calculation.

Estimation of plutonium inhalation uptake per day

Lawrence Livermore National Laboratory (LLNL) has been monitoring the exposure of the Marshallese through environmental methods (Noshkin et al. 1979, 1981, 1988, 1994; Robison et al. 1980, 1982, 1987, 1988; Robison 1983; Robison and Stone 1992). The LLNL group analyzed plutonium concentrations in air, soil, water, and food. These data, combined with the observed dietary patterns in the Northern RMI, are then used to estimate plutonium intake and committed doses. On the other hand, the BNL group analyzed plutonium concentration in 24-h urine samples to estimate plutonium uptake and committed dose (Lessard et al. 1984; Sun et al. 1993a, 1995). An intake model developed and used by the LLNL group for assessing plutonium dose via inhalation pathway for Marshallese based on their environmental and the life style conditions estimated a total activity intake of about $180 \mu\text{Bq d}^{-1}$ for both ^{239}Pu and ^{240}Pu . This is based on the product of $22 \text{ m}^3 \text{ d}^{-1}$ breathing rate and the $^{239+240}\text{Pu}$ concentration $8 \mu\text{Bq m}^{-3}$ in the air breathing zone (Robison et al. 1987, 1982, 1989). A similar value was also reported by Kohn (1989) for assessing potential inhalation intake of plutonium for the people of Rongelap.

Because ^{240}Pu is not measured by the FTA method, the relative proportions of ^{239}Pu to ^{240}Pu must be estimated if the FTA measurement results are to be compared with the LLNL results. Oak Ridge National Laboratory reported a ^{239}Pu to ^{240}Pu activity ratio of 20:27 for another thermonuclear device detonated on Enewetak in 1952, code name Ivy Mike (Holleman et al. 1987). Since the plutonium compositions produced by the Bravo and Ivy Mike devices were similar, the present work applies the Ivy Mike ratio to obtain the LLNL inhalation value of $77 \mu\text{Bq d}^{-1}$ for ^{239}Pu .

ICRP Publication 48 (1986) reported a range of pulmonary clearance rate of 0.0002 to 0.0010 d^{-1} for all $^{239}\text{PuO}_2$ and $^{239}\text{PuO}_2\text{:UO}_2$ depending upon the particles'

sizes and solubility in body fluids. ICRP Publication 48 also indicates that the rate of lung clearance could be even slower than 0.0002 d^{-1} for plutonium compounds fired at high temperatures (i.e., over $1,000^\circ\text{C}$). This is a valid concern in estimating the plutonium burdens in the Marshallese, since the plutonium remaining on the islands primarily is an insoluble, high-fired oxide (Schell and Walters 1975; Noshkin et al. 1981). A recent study on pulmonary clearance for workers with chronic exposure to Class Y uranium indicated that the rate of the lung clearance is about 0.0001 d^{-1} (Dang et al. 1994). For various oxide forms of Type S (corresponding to "Class Y") plutonium particles (assume $1\text{--}5 \mu\text{m-AMAD}$), the ICRP's Human Respiratory Tract Model (HRTM) (1994) indicated that about 10% of inhaled particles cannot be exhaled. The activity associated with these 10% particles is eventually deposited in the alveolar-interstitial (AI) compartment, the pulmonary region of the lung. About 10% of the AI-deposited plutonium reaches the blood. The HRTM (ICRP 1994) also indicates that 0.1% of the AI deposited plutonium can be rapidly absorbed to blood and affects the interpretation of urine measurements.

Using both the estimates of $77 \mu\text{Bq d}^{-1}$ inhalation intake rate for ^{239}Pu and the HRTM default value 10% for inhaled activity deposited to the pulmonary region of lung, the calculated deep lung deposition rate is $7.7 \mu\text{Bq d}^{-1}$. Because of both 0.1% of instantaneously rapid absorption and 1% of the cumulative absorption from AI compartment to systemic blood, the estimated total plutonium uptake via lung (inhalation pathway) is $0.077 \mu\text{Bq d}^{-1}$ [i.e., $7.7 \mu\text{Bq d}^{-1} \times (0.001+0.01)$]. This contribution is small and can be negligible relative to the total intake at $14.2 \mu\text{Bq d}^{-1}$. Hence, the uptake contribution due to the potential inhalation pathway is not considered further.

Estimation of plutonium ingestion intake per day

Although the dietary pattern is important when estimating ingestion uptake, particularly for populations such as the Marshallese, other aspects of their life style also must be considered. For example, sleeping on the floor, preparing food outdoors, and eating in their often dusty environments increases the possibility that deposited plutonium will enter the body through ingestion (Lessard et al. 1985; Simon 1994; Baverstock et al. 1995). Table 2 is an abridged version of the LLNL dietary pattern for the Rongelap Islands [all dietary items contributing less than $37 \mu\text{Bq d}^{-1}$ have been omitted (Robison et al. 1989)]. Based on Table 2, the total dietary intake of ^{239}Pu and ^{240}Pu is about 5 mBq d^{-1} (Robison et al. 1989). Using the Ivy Mike ratio of 20:27 for ^{239}Pu : ^{240}Pu , the dietary intake of ^{239}Pu alone would be about 20 mBq d^{-1} . Since the plutonium (both ^{239}Pu and ^{240}Pu) concentration in Rongelap soil is reported (Robison et al. 1989) to be 150 mBq g^{-1} (4 pCi g^{-1}), about 64 mBq g^{-1} would be ^{239}Pu . Similarly, Baverstock et al. (1995) indicates the average and standard deviation of $^{239+240}\text{Pu}$ concentration in 0–5 cm topsoil (8 samples) at the Rongelap Island were $198 \pm 140 \text{ mBq g}^{-1}$ with a

Table 2. An abridged version of the model Marshallese diet from Robison et al. (1989). All dietary items contributing less than 37 $\mu\text{Bq d}^{-1}$ have been omitted.

Local food	Ingestion rate g d^{-1}	Intake rate Bq d^{-1} ($^{239,240}\text{Pu}$)
Reef fish	24.20	2.1×10^{-4}
Tuna	13.90	1.2×10^{-4}
Marine crabs	1.68	6.7×10^{-5}
Lobster	3.88	1.6×10^{-4}
Clams	4.56	1.7×10^{-3}
Trochu ^a	0.10	3.7×10^{-5}
Tridacna Muscle ^a	1.67	6.3×10^{-5}
Jedrul ^a	3.08	1.2×10^{-3}
Coconut crabs	3.13	2.3×10^{-4}
Octopus	4.51	4.4×10^{-5}
Chicken liver	4.50	5.6×10^{-5}
Pork liver	2.60	8.9×10^{-5}
Coconut juice	99.10	9.6×10^{-5}
Coconut milk	51.90	8.5×10^{-5}
Drinking coco meat	31.70	4.1×10^{-5}
Arrowroot	3.93	1.0×10^{-4}
Well water	207.00	1.0×10^{-4}

^a Clam or shellfish-related species.

median value of 155 mBq g^{-1} . The uncertainty of Rongelap's soil measurement is estimated to be about 0.7 at 67% confidence level.

Estimation of plutonium intake from soil consumption

Harrison et al. (1989) and Haywood and Smith (1990) reported an average soil intake of 10,000 mg d^{-1} in dose assessments for the Emu and Maralinga nuclear weapons testing sites in Australia. As a result of resuspension, soil dust can be deposited on plants and in food during preparation. Hence, it may be ingested directly. A soil ingestion rate of 100 mg d^{-1} as a default was chosen for a pathway analysis for the U.S. population (Yu et al. 1993). The assessments included inhaling suspended dust, drinking water, and ingesting food contaminated with deposited dust. Haywood and Smith specifically discussed the effects of lifestyle on plutonium ingestion for the Australian aboriginal people; an average soil intake of 1,000 mg d^{-1} was established from the fecal samples of the investigators who made field trips to the affected areas. Therefore, the 1,000 mg d^{-1} soil intake is regarded by Haywood and Smith as the lowest limit on soil intake for the aboriginal people, and it is in general agreement with populations exhibiting habitual pica (the deliberate ingestion of soil) (Schaum 1984; LaGoy 1987). A health risk impact study of the U.S. population also suggested that children who eat soil can ingest as much as 5,000 mg d^{-1} directly without exhibiting ill effects (Cleverly 1987).

It is difficult to quantitatively compare the amount of soil ingested by the Marshall Islanders and the Aboriginal people because of their different lifestyles. However, both societies live in close contact with their natural environment, although the Australian aboriginal people are nomadic, while the Marshallese have a life-

style more nearly like to that of industrial nations. LaGoy (1987) reported a maximum intake of 500 mg d^{-1} for adults in developed nations who do not exhibit habitual pica. This value, then, was taken to be a reasonably conservative average for the Marshallese people. Therefore, this work adopts 500 mg d^{-1} as the average life-time intake of soil by the Marshallese (Table 3).

RESULTS AND DISCUSSION

Derivation of f_1 value for Marshallese

Since the fraction of the ingested plutonium absorbed that is reaching the systemic system is the f_1 value, we can estimate f_1 by dividing the daily uptake to blood of 14.2 $\mu\text{Bq d}^{-1}$ by the total intake rate. Using the 500 mg d^{-1} soil ingestion rate value, the ^{239}Pu intake is estimated to be 0.032 Bq d^{-1} . Combining this with an estimate of dietary intake of 0.002 Bq d^{-1} gives a total intake of 0.034 Bq d^{-1} . The estimated f_1 value is

$$f_1 = \frac{14.2 \times 10^{-6}}{0.034} \approx 4.2 \times 10^{-4}. \quad (1)$$

This f_1 value is about the same as we presented earlier (Sun et al. 1993b). Outlines of the calculation and parameter values used are summarized in Table 4. Using the median value of 0.6 $\mu\text{Bq d}^{-1}$ for uptake, an f_1 value of 3.8×10^{-4} is calculated instead. Sensitivity analyses between f_1 values vs. soil ingestion rates were performed, with results tabulated in Table 5. The maximum estimated f_1 value could be as high as about 7×10^{-3} . Also, the plutonium intake rate from ingested soil would equal that of dietary sources at about 30 mg d^{-1} soil ingestion rate. Above this value, plutonium intake from soils exceeds that from dietary intake.

Among the 34 participants whose urine results were used in this study, two are among the original 64 inhabitants who evacuated Rongelap in 1954. Unlike the others, these two individuals both observed the fallout, went through the 3 y of exile, and then lived a full 31 y on Rongelap Island after returning. However, both had less than 3.7 μBq in their 24-h urine samples or a committed effective dose about 0.37 mSv to age 70 y (Sun et al. 1995). This suggests that the impact on internal exposure of plutonium during the direct fallout in March 1954 was not significant. A similar conclusion

Table 3. Estimates of soil ingestion rates.

Study year	Estimate (mg d^{-1})		Applicable population
Schaum (1984)	100	Avg.	2–6 y, U.S.
	5,000	Max.	2–6 y, U.S., Habitual pica
LaGoy (1987)	500	Max.	Adult, U.S.
	5,000	Max.	Adult, U.S., Habitual pica
Haywood and Smith (1990)	1,000	Avg.	Adult, Aborigine
Robison et al. (1989)	10	Avg.	Adult, Marshallese Based on dietary intake
This work	500	Avg.	A lifetime average, Marshallese

Table 4. Outline of the f_1 value calculation with assumptions and related parameter values.

Intake estimates		The ^{239}Pu and ^{240}Pu activity ratio in the Bravo dust was taken to be 20:27.
A: Inhalation pathway		= $77 \mu\text{Bq d}^{-1}$ (The products of $22 \text{ m}^3 \text{ d}^{-1}$ breathing rate times the plutonium concentration of $8 \mu\text{Bq m}^{-3}$ times the ^{239}Pu and ^{240}Pu activity ratio). 10% of the inhaled plutonium particles are assumed to be deposited in the pulmonary region (AI). Hence, the deposited ^{239}Pu in lung is $7.7 \mu\text{Bq d}^{-1}$.
B: Ingestion pathway		
1. Via dietary intake		= 0.002 Bq d^{-1} (The products of $5 \mu\text{Bq d}^{-1}$ diet intake rate times the $^{239,240}\text{Pu}$ concentration of $8 \mu\text{Bq m}^{-3}$ times the ^{239}Pu and ^{240}Pu activity ratio.)
2. Via ingested soil		= 0.032 Bq d^{-1} (The products of 500 mg d^{-1} intake rate times the soil concentration of 150 mBq g^{-1} times the ^{239}Pu and ^{240}Pu activity ratio.)
Uptake Estimates		
A: Via inhalation pathway		= $(7.7 \mu\text{Bq d}^{-1}) \times (0.001 + 0.01) = 0.077 \mu\text{Bq d}^{-1}$, the fractional transferred to blood for Type S particles are 0.1% and 1% for instantaneously and cumulative absorptions, respectively (ICRP 1994).
B: Via ingestion pathway		= $[(0.032 + 0.002) \text{ Bq d}^{-1}] \times f_1^a$ (dietary plus ingested estimate given above).
C: Via 24-h FTA urinalysis		= $14.2 \mu\text{Bq d}^{-1}$.

$^a f_1$ = urinalysis result - the uptake to blood via inhalation divided by the total ingestion activity. Therefore, $f_1 = (14.2 - 0.077) \mu\text{Bq d}^{-1} \div [(0.032 + 0.002) \text{ Bq d}^{-1}] = 4.2 \times 10^{-4}$.

Table 5. Sensitivity analysis for f_1 value due to various soil ingestion rates for Marshallese.

Soil intake rate (mg d^{-1})	Plutonium intake from soil (Bq d^{-1})	Plutonium intake from diet (Bq d^{-1})	Total intake (Bq d^{-1})	f_1 Value
0		2.0×10^{-3}	2.0×10^{-3}	7.1×10^{-3}
10	6.4×10^{-4}	2.0×10^{-3}	2.6×10^{-3}	5.4×10^{-3}
30	1.9×10^{-3}	2.0×10^{-3}	3.9×10^{-3}	3.6×10^{-3}
50	3.2×10^{-3}	2.0×10^{-3}	5.2×10^{-3}	2.7×10^{-3}
100	6.4×10^{-3}	2.0×10^{-3}	8.4×10^{-3}	1.7×10^{-3}
200	1.3×10^{-2}	2.0×10^{-3}	1.5×10^{-2}	9.6×10^{-4}
300	1.9×10^{-2}	2.0×10^{-3}	2.1×10^{-2}	6.7×10^{-4}
500	3.2×10^{-2}	2.0×10^{-3}	3.4×10^{-2}	4.2×10^{-4}
1,000	6.4×10^{-2}	2.0×10^{-3}	6.6×10^{-2}	2.2×10^{-4}

was drawn from a study of bone samples of deceased Rongelap peoples who were on Rongelap Island (Franke et al. 1995). Therefore, all the estimated f_1 values presented in this study represent chronic exposure of the people of Rongelap to a plutonium-contaminated environment.

Influence of factors other than soil ingestion on estimates of f_1 values

Although the major assumption in this paper is that increasing the assumed soil ingestion rate thereby reduces the deduced f_1 value, other parameters which might also reduce the estimated uptake have been conservatively evaluated.

First, there was no allowance for the assumption that the specific activity associated with airborne plutonium is 2.5 times greater than that for the soil (Shinn et al. 1980); however, the top layer of soil can be expected to reflect the specific activity of that in the air. Then, it might be assumed that the ingested soil has a specific activity 2.5 times that of the soil averaged over 5 cm depth; such an assumption would lower the f_1 value by a factor of about 2. Second, there was no allowance for

plutonium transfer during embryonic development following intake by the mother, which also would somewhat lower estimates of the value ($\sim 10\%$) (Morgan et al. 1992; Stather et al. 1992). Third, no allowance was made for a large f_1 value (about 10 times higher) during infancy. Since this enhanced uptake occurs over a relatively short period of life, the effect would be to lower the estimated f_1 value by a factor of 1.5. Fourth, there also was no allowance for intake through open skin cuts and wounds (Geiger and Sanders 1956; Piechowski et al. 1989). Such intakes could enhance uptake, thereby reducing the estimate of the f_1 value.

Marshall Islands soil was created from coral reef. Hence, the basic topsoil components in the Rongelap Atoll consist of sand, small fragments of marine shells, and hard corals. Therefore, plutonium in the soil samples from the Marshall Islands exists with lime enriched compounds.) Laboratory experimentation shows that bulk soil from the Marshall Islands can be easily dissolved in weak acids similar to those encountered during digestion (Simon[†]). It is likely that the f_1 value is governed both by the oxidation states of the plutonium and the compounds it binds with during digestion. This may explain why the f_1 value calculated for the people of Rongelap Island from plutonium oxide is about double the ICRP (1995) recommended value ($f_1 = 10^{-5}$).

CONCLUSIONS

The objectives of this study were to provide a method using urine data for assessing GI tract absorption constant and to determine an appropriate value for the Marshallese populations. All values for critical parameters for estimating the f_1 value for the Marshallese are based on environmental concentrations of plutonium in the Rongelap Islands and in low-level plutonium urinal-

[†] Simon, S. L. Private communication, National Academy of Sciences, 2101 Constitution Ave, N. W., Washington, DC 20418 1995.

yses. In addition to the uncertainty associated with the methodology and measurements used in this assessment, there are other parameters that could influence the determination of the f_1 value. Because of both the Marshallese life style and the levels of plutonium present in the topsoil, we must emphasize the importance of estimating the most realistic soil ingestion intake rate for assessing the f_1 value. Overestimating the rate of intake could result in a fivefold overestimation of total intake for some individuals. Table 5 also shows that the most sensitive factor for estimating f_1 value is the soil ingestion rate (i.e., more important than dust resuspension, embryo uptake from the mother, and open skin absorption). The 500 mg d⁻¹ soil ingestion rate is an assumed single value. Increasing the soil ingestion rate will decrease the f_1 value, and visa versa. Using both parametric and non-parametric statistics, 4×10^{-4} is found as a realistic f_1 value for the Marshallese.

The f_1 value of 4×10^{-4} calculated for adults is applicable to children as well. This value is in substantial agreement with studies in which plutonium was chronically fed to young animals (NRPB 1990) and with the values recommended for children ages 1 y and older (ICRP 1993). We conclude that the f_1 value of 5×10^{-4} recommended by the ICRP (1993, 1995) is the most appropriate one for assessing plutonium exposure to the Marshall Islands population.

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